

RemarksApplication Status and Disposition of Claims

This is in response to the non-final Office Action mailed December 31, 2009. Inasmuch as the three-month shortened statutory period is originally set in the Office Action to expire on March 31, 2010, Applicant files this response concurrently with a Request for Two-Month Extension of Time and required fee.

With this amendment, no claims are amended or canceled, and new claim 21 is added. This amendment finds support throughout the specification, and particularly, for example, at page 6, paragraph [0091] of the published specification. The amendment adds no new matter.

Claim Rejections – 35 U.S.C. § 103(a)

The Office Action rejects claims 1, 3, 9-12, 14, 15, 19, and 20 under 35 U.S.C. § 103(a) as being obvious over Maekawa et al. (WO 2002/090435) in view of Nakano et al. (U.S. Patent No. 5,302,645) and Kudou et al. (US 2002/0123570). The Action rejects claims 12 and 13 under 35 U.S.C. § 103(a) as being obvious over Maekawa et al. in view of Nakano et al., Kudou et al., and Largman et al. (US 4,403,052), with evidence provided by MatWeb (ENC 1299). Applicants respectfully disagree with the rejections for at least the following reasons.

Applicants note that both rejections rely on Maekawa et al. as the primary reference, with secondary teachings provided by Nakano et al. (and others). The Office Action states that Maekawa et al. does not specifically discuss the content of the epoxy resin or polycarbonate resin or the particular use of the resin product. (Page 3, last sentence of first full paragraph.) For this missing teaching, the Office Action relies on Nakano et al. (See page 3, second paragraph.) The sole basis for the Office Action's reliance on Nakano et al. to provide the features of Applicants' claimed invention – that the Office admits are missing from Maekawa et al. – is a single sentence: "Nakano teaches a polyethylene terephthalate composition, *which is a similar alkylene terephthalate resin.*" Applicants respectfully note that this assertion is completely unsupported

in the record, and thus, insufficient to support the obviousness rejection. And more importantly, it is factually incorrect.

Initially, with respect to the support for the assertion that polytrimethylene terephthalate (PTT) is “similar” to polyethylene terephthalate (PET), Applicants note that the Office had previously made the argument in an Office Action dated May 28, 2008 that the claimed invention would be obvious over Nakano et al. and that it would be obvious to substitute the claimed PTT for Nakano et al.’s PET, given the *advantages of improved mechanical properties* of PTT over PET described in other references – Houck and Nexant. (Applicants note that that rejection was ultimately withdrawn.) Applicants note that the basis for that rejection was not that the two polyester resins were *similar*, but rather, that PTT was better than PET and that a person skilled in the art would substitute PTT for PET in an effort to obtain *different and improved* characteristics of PTT over PET. Indeed, Applicants respectfully submit that Houck and Nexant do not stand for the proposition that PET and PTT are “similar”; rather, Houck and Nexant explain why PTT has advantages over PET, as well as polybutylene terephthalate (PBT).

Applicants respectfully submit that Houck and Nexant discuss similarities of PET, PBT, and PTT, but mostly focus on the differences (and advantages of PTT). Applicants note that Houck and Nexant discuss how these three commercially important polyester resins are structurally similar, and how their synthesis is similar. However, simply because the three resins are all polyesters formed by similar condensation reactions does not mean that a person skilled in the art would consider them “similar” for purposes of combining the reference teachings. Applicants submit that a person skilled in the art recognizes that the structural differences result in different physical properties and characteristics, which lead to different particular end uses.

For example, Houck specifically notes that PTT has an odd number (three) of methylene unites between each of the terephthalates, whereas PET and PBT have an even number, which gives the PTT an elastic recovery beyond that of PBT or PET. Houck also notes that PTT is advantageous in its dyeability. Nexant notes the difference in the elasticity (see page 3) – noting that PTT has nearly 100% elastic recovery, whereas PET only has 75-80%. Nexant also notes that the melting temperature of PET is 265°C, whereas PTT is only 225°C, which is a significant

difference. Also, the glass transition temperature (as noted in Nexant) is 45-65°C for PTT and 80°C for PET.

Applicants respectfully refer the Office to the attached publication from Shell Chemicals, which describes PTT and shows how it differs from PBT and PET. In particular, Applicants refer the Office to Figure 9, which shows the very different elastic recovery profiles of PET and PTT. Applicants also specifically refer the Office to Figure 14, which shows the different impact resistance (Notched Izod Impact) values for PET, PBT, and PTT. Applicants further refer the Office to Figure 15, which graphically demonstrates the differences in tensile properties between PTT and PET. Figure 16 shows the very different profiles for shrinkage force v. temperature for PTT compared to PET, and Figure 17 shows percent shrinkage v. temperature for PTT compared to PET. Applicants respectfully submit that the characteristics shown in these figures would be important in considering the use of a particular polyester in a molded article for a bath product, washroom product, toilet product, kitchen product, and sink product.

In view of the foregoing, Applicants respectfully submit that PET and PTT would not be considered "similar" by a person of ordinary skill in the art, and would not have combined the teachings of Maekawa et al. and Nakano et al. as the Office has. Applicants respectfully submit that the rejections under 35 U.S.C. § 103, both of which rely on the combination of Maekawa et al. and Nakano et al., are improper for at least this reason, and should be withdrawn.

Applicants note that the Office further relies on Kudou et al. for its disclosure of a resin composition having good mechanical properties such as impact resistance and heat stability, in addition to good surface appearance, obtained by the use of inorganic fillers such as wollastonite. However, Applicants note that Kudou et al. relates to a *polyoxymethylene resin composition* comprising 40 to 99.5 parts by weight polyoxymethylene resin (A) and 0.5 to 60 parts by weight consisting of a polyamide elastomer (B) and an acid-modified olefinic resin (C), which is entirely different from the polytrimethylene terephthalate resin composition of the present invention. In addition, Kudou et al. teaches that inorganic fillers can be used to impart desired surface appearance and slidability to molded articles (paragraph [0063]). Applicants respectfully note that Kudou et al. is silent with respect to the use of wollastonite to provide mechanical strength,

which is the purpose for which Applicants include the wollastonite (in addition to appearance). More importantly, however, Applicants submit that a person skilled in the art would not combine the teachings of Kudou et al., which relate to polyoxymethylene resin compositions, with either of Maekawa et al. or Nakano et al., which relate to completely different polymer systems.

Finally, for the sake of completeness, Applicants note that new claim 21 recites that the claimed molded article has a Barcol hardness of 40 or more. Applicants respectfully submit that the cited art, alone or in combination, fails to render obvious this feature of the presently claimed invention. At the very least, Applicants submit that (in addition to the points raised above) there is no expectation of success in achieving a Barcol hardness of 40 or more based on the cited art. Thus, Applicants submit that not only does the cited art not suggest that one *should* make a molded article with the claimed features having a Barcol hardness of 40 or more, the cited art does not suggest that one *could* make such a molded article. Applicants submit that the rejection should be withdrawn for at least this reason as well.

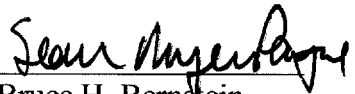
In view of the foregoing remarks and amendments, Applicant respectfully submits that the cited art, taken in any proper combination, fails to render obvious the presently claimed invention. Applicant respectfully requests withdrawal of the obviousness rejections.

Conclusion

In view of the foregoing, it is believed that all of the claims in this application are in condition for allowance, which action is respectfully requested. If any issues yet remain which can be resolved by a telephone conference, the Examiner is respectfully invited to telephone the undersigned at the telephone number below.

Should the Examiner have any questions, the Examiner is invited to contact the undersigned at the below-listed telephone number.

Respectfully submitted,
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Enclosure: "Poly(trimethylene phthalates or naphthalate) and Copolymers:
New Opportunities in Film and Packaging Applications,"
Hwo et al., presented at the Future-Pak 98 Conference, Chicago,
November 10-12, 1998

May 28, 2010
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Shell Chemicals

Poly(trimethylene phthalates or naphthalate) and Copolymers: New Opportunities in Film and Packaging Applications.

Charles Hwo, Tom Forschner, Rohan Lowtan, Dave Gwyn, Barry Cristea
Presented at the Future-Pak® 98 Conference, November 10-12 1998, Chicago

Abstract

Poly(trimethylene phthalates or naphthalate) (PTT, PTI, PTN) and copolymers are aromatic polyesters made by polycondensation of 1,3-propanediol (PDO) and terephthalic acid, isophthalic acid or naphthalic acid and/or with comonomers (isophthalic acid, 1,4-butanediol etc.). This brochure describes the chemistry and properties of such PDO-based polyesters and their applications in films and packaging. Key performance properties of the several PDO-based polyesters including poly(trimethylene naphthalate) will be compared with poly(ethylene terephthalate), poly(ethylene naphthalate) and nylon 6.

Introduction

Poly(trimethylene terephthalate) (PTT) has been recently introduced as a commercial polymer, joining the other aromatic polyesters, poly(ethylene terephthalate) (PET) and poly(butylene terephthalate) (PBT). Commercialization was made viable by a breakthrough in synthesis of the monomer 1,3-propanediol (trimethylene glycol) (Fig. 1) which enabled the polymer to be produced at costs suitable for commercialization. PTT is generally made by the melt condensation polymerization of 1,3-propanediol (PDO) and terephthalic acid (Fig. 2). The polymer produced is similar in characteristics such as molecular weight and molecular weight distribution to other polyesters (Fig. 3) and is intended for fiber, film, and Engineering Thermoplastics (ETP) markets, particularly those dominated by nylons, PET, or PBT.

Although the synthesis of PTT is not new, the developing technology surrounding this polymer appears to offer the promise of opportunities for uses of this polymer in the carpet (1), textile (2), film and packaging (3), ETP (4) and other market-

places (5). Details of the technology development related to the PDO-based polyesters (PTT and its copolymers, poly(trimethylene naphthalate) (PTN), poly(trimethylene isophthalate) (PTI)) in film and packaging end uses or applications will be shared in this brochure.

Physical characteristics

PTT is a rapidly crystallizing linear aromatic polyester with a crystal melting temperature of 225 °C and a glass transition temperature of about 50 °C (Fig. 4). The semi-crystalline solid is an opaque rigid thermoplastic useful for structural ETP applications where rigidity, strength and toughness are required. The molten polymer can be extruded into sheet and biaxially stretched into films. The chain conformation of the odd-numbered *n*-methylene glycol derived polymer results in a unique crystal structure (Fig. 5), which is very "spring-like" in its long axis (Fig. 6), and results in high resilience (Figures 7, 8 and 9). PTT films exhibit softness and good high shrinkage and shrink force at low temperatures. Other 1,3-propanediol based homopolymers and copolymers exhibit unique basic properties, which may improve the processing of PTT, and also offer desired performance properties.

Rheological characteristics

The melt rheology of PTT is very similar to that of PBT. The viscosity shear rate relationship (Fig. 10) goes from moderately non-Newtonian at temperatures 10-15 °C above T_m to quite Newtonian at temperatures of 40 to 50 °C above T_m . A typical grade of PTT suitable for fiber-spinning has a melt viscosity, (η_a), of about 200 Pa-sec at 260 °C and at shear rate of 200 1/sec (Fig. 11). The melt rheology of PTT polymers truly reflects the molecular characteristics of the polymer. Average molecular

weight, molecular weight distribution, skewness, entanglement length and degradation by hydrolytic cleavage are all classically expressed in the zero shear viscosity, the power law index, the die swell ratio, the normal force or melt elasticity and the rate of change of viscosity with time.

Processing characteristics

PTT polymers can be processed by typical thermoplastic melt processes, extrusion, injection molding, blow molding and melt-spinning, to name a few. The molten polymer has good melt strength and can be easily extruded into films. Like all polyester polymers, careful attention must be given to proper drying of the polymer nibs prior to any melt forming process to prevent molecular weight degradation by hydrolysis (Fig. 12). Typical processing temperatures are about 250 °C to 265 °C for typical thermoplastic processes and film extrusion. To make oriented film, the stretching temperature is much lower and the stretching temperature window is much narrower than those of PET (Fig. 13) are. High line speed is preferred.

Mechanical, electrical, chemical resistance properties of PTT compared to other ETP polymers

The mechanical and electrical properties of "neat" injection molded PTT polymer are intermediate between those of PBT and those of PET (Fig. 14). This includes flexural modulus, HDT at 264 psi and notched Izod impact. Properties, which are much closer to those of PET, include tensile strength and dielectric strength. On the whole, the mechanical properties of the polyester family are inferior to those of nylon 6 and nylon 6,6, while electrical properties are superior. The only other significant polymer with use temperature in the range of the polyesters is amorphous polycarbonate, which is greatly superior in both HDT and impact strength but inferior in all other mechanical and electrical properties. PTT polymers share with the polyesters an inherent resistance to organic solvents and foods as well as to inorganic acids and bleaches.

Film properties

Mechanical properties

Comparison of biaxially oriented film properties of PTT, PET and nylon 6 indicates that PTT is significantly softer (Fig. 15) than PET which is similar to observations in carpets and textiles. PTT films also show higher shrink force (Fig. 16) and percent shrinkage (Fig. 17) than either PET or nylon 6. This means that PTT will require lower shrink tempera-

ture than PET for the same shrink force or percent shrinkage.

Gas transmission rates

Both of the developmental PDO-based polyesters: poly(trimethylene naphthalate) (PTN) and poly(trimethylene isophthalate) (PTI) exhibit excellent barrier effects to carbon dioxide (Fig. 18), water vapor (Fig. 19) and oxygen (Fig. 20) in comparison to PET and nylon 6. This suggests that an all polyester-based high-barrier packaging is possible by using the developmental PDO based polyesters, PTN or PTI to replace polyvinylidene chloride (Saran) or ethylene-vinyl alcohol copolymer (EVOH). In doing so, the potential for recycling of the PTN or PTI laminated PET film or container (bottle, tray, etc.) is enhanced.

Heat sealability

The developmental PDO-based polyester, PTI can be heat sealed at low temperature (Fig. 21) like polyethylene and PVDC. Unlike the polyethylene seal coat, a tie layer adhesive is not required for using PTI as heat seal coat for PET. Therefore, a heat sealable all polyester film is then possible and again the potential for recycling of the PTI coated PET film or tray is enhanced.

Acknowledgement

The following colleagues have also contributed the work at some point of time: J. Grebowicz, B. M. Scardino and Frank Smith.

References

1. H. H. Chuah, CORTERRA® Poly(trimethylene terephthalate) - *New Polymeric Fiber for Carpets*, Chemical Fibers International, Vol. 46, pp. 424-428, 1996.
2. H. S. Brown, H. H. Chuah, *Texturing of Textile Filament Yarns Based on Poly(trimethylene terephthalate)*, Chemical Fibers International, Vol. 47, pp. 72-74, 1997.
3. Charles Hwo, Kailash Dangayach, Tom Forschner, Barry Cristea, Frank Smith, Dave Gwyn, Kalyan Ghosh, Houston Brown, *Poly(trimethylene phthalates or naphthalate) and Copolymers: New Opportunities in Film, Engineering Thermoplastics and Other Applications*, 1998 Science, Engineering and Technology Seminars, May 23-25, 1998, Adams Mark Hotel, Houston, Texas.
4. Kailash Dangayach, Hoe Chuah, William Gergen, Phil Dalton and Frank Smith, *Proceedings of the SPE 55th Annual Technical Conference*, pp. 2097-2101, 1998.

5. Houston S. Brown, Paul K. Casey and J. Mike Donahue, *Poly(trimethylene terephthalate)* *Polymer for Fibers*, Shell Chemical Company Publication No. SC:2599-97, 10, 1997.

Figure 1/Synthetic Routes to PDO.

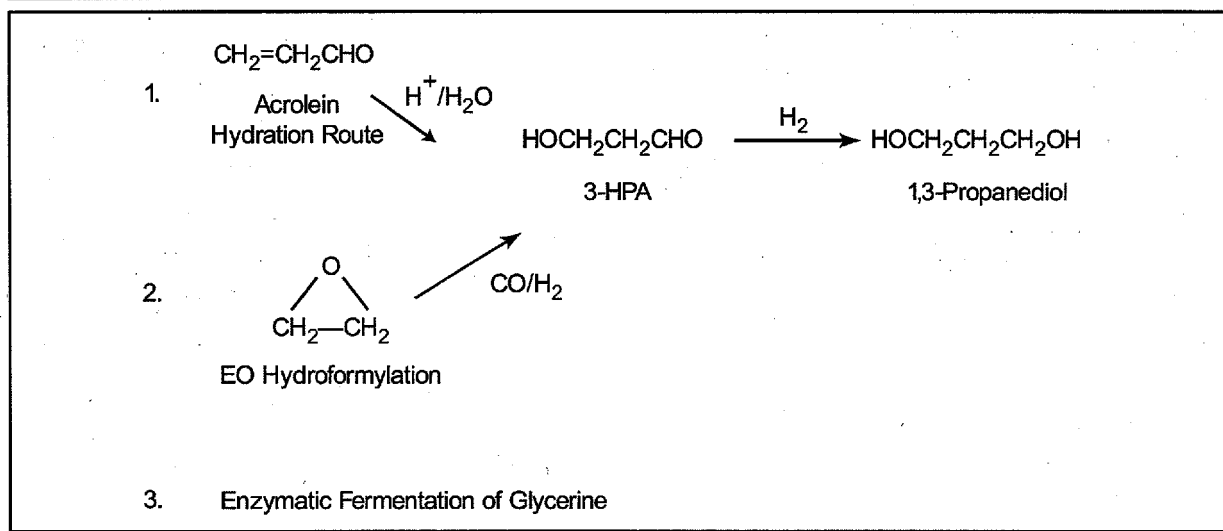


Figure 2/TPA Process.

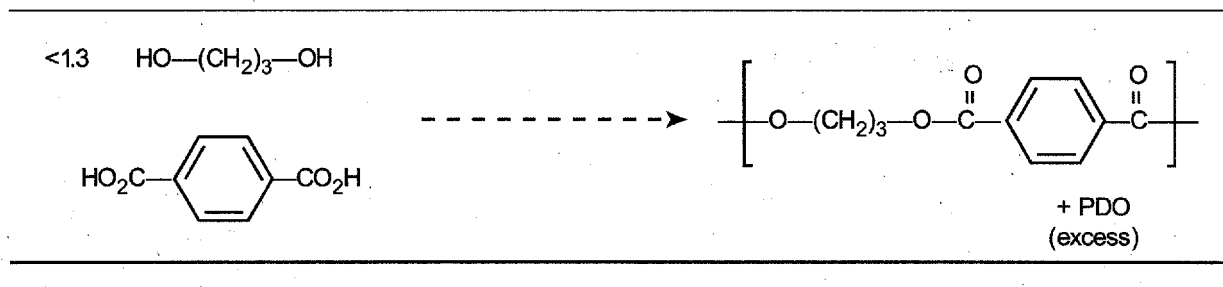


Figure 3/Family of Commercial Aromatic Polyesters.

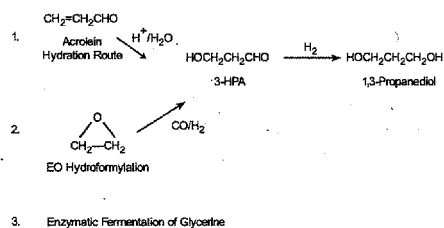


Figure 4/Basic Physical Properties of ETP Polymers.

Physical Property	PET	PTT	PBT	Nyl 6,6	PC	Nyl 6
Specific Gravity	1.40	1.35	1.34	1.14	1.20	1.14
Melt Temperature, °C	265	225	228	265		230
Glass Trans. Temp., T_g , °C	80	45-75	25	50-90	150	50

• PTT data are reported from measurements made in our laboratory.
 • All other data are taken from the available polymer literature.

Figure 5/Crystallographic Data.

PET, PTT & PBT Are Triclinic

	PET ¹	PTT ²	PBT ³
a, Å	4.56	4.6	4.83
b	5.94	6.2	5.95
c	10.75	18.3	11.67
α , °	98.5	98	100
β	118	90	64
γ	112	112	69
Crystal Density (g/cm ³)	1.515	1.387	1.395
% Chemical Repeat Length	99.5	75.3	86.3

1. Daubeny R.D.P. *et al.*, *Proc. Roy. Soc. (London)*, A226, 531 (1954).
2. Desborough. I.J., *Polymer*, 20, 545 (1979).
3. Boyle, C.A., *Bull. Am. Phys. Soc.*, 19, 352 (1974).

Figure 6/Confirmation of Polyester Polymers in the Crystalline Phase.

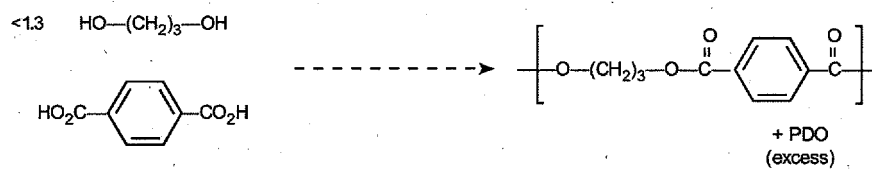


Figure 7/CORTERRA™ Polymer PTT Elastic Recovery—Oriented.

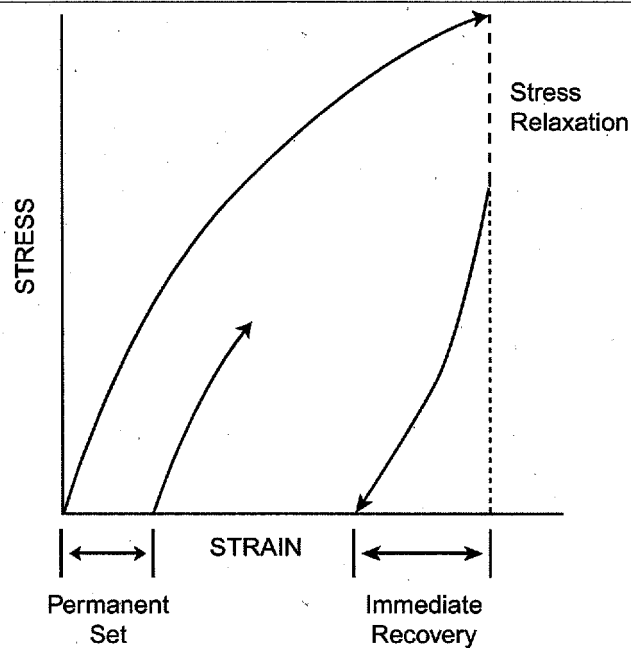


Figure 8/Elastic recovery of PTT, PET and Nylon 6,6 Immediate Recovery.

PET, PTT & PBT Are Triclinic

	PET ¹	PTT ²	PBT ³
a, Å	4.56	4.6	4.83
b	5.94	6.2	5.95
c	10.75	18.3	11.67
α, °	98.5	98	100
β	118	90	64
γ	112	112	69
Crystal Density (g/cm ³)	1.515	1.387	1.395
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2. Desborough. I.J., *Polymer*, 20, 545 (1979).

3. Boyle, C.A., *Bull. Am. Phys. Soc.*, 19, 352 (1974).

Figure 9/Elastic Recovery of PTT, PET and Nylon 6,6 Permanent Set.

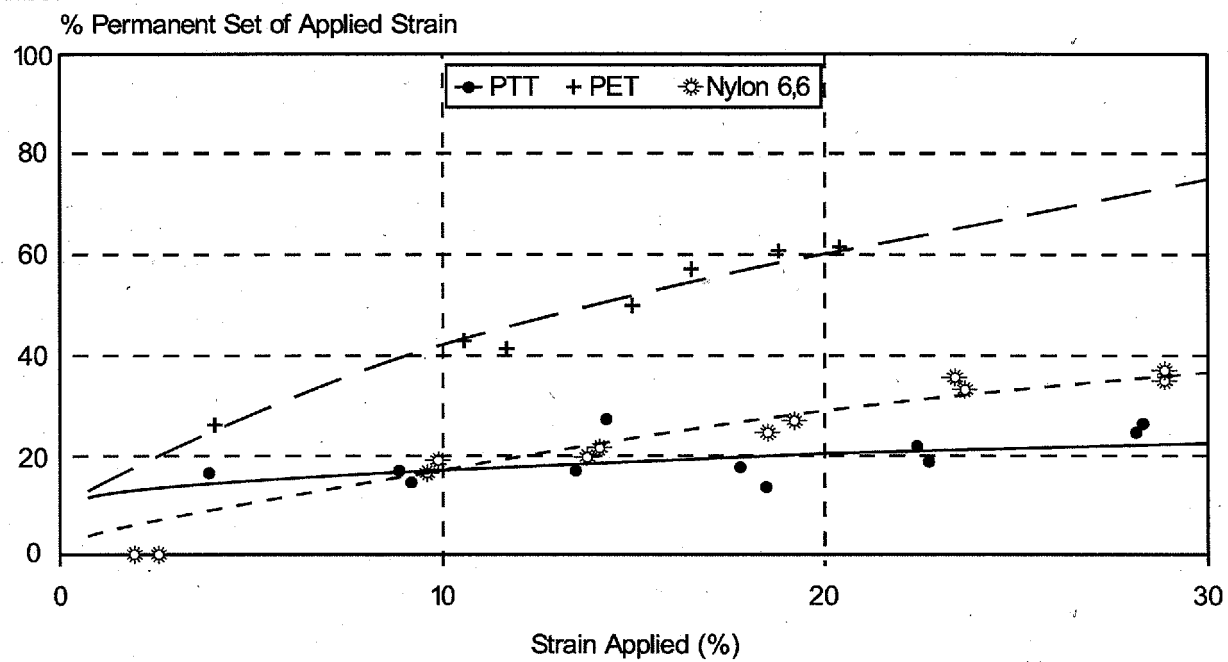


Figure 10/Melt Viscosity Power Law Exponents.

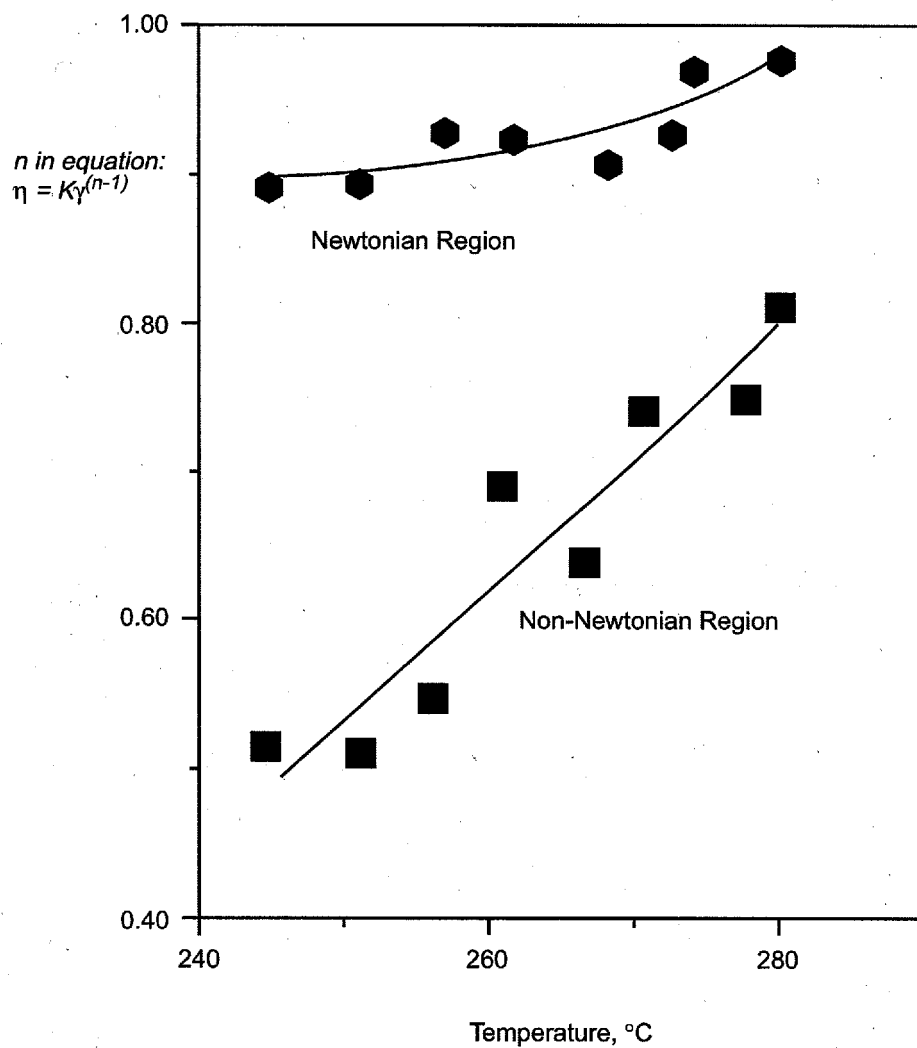
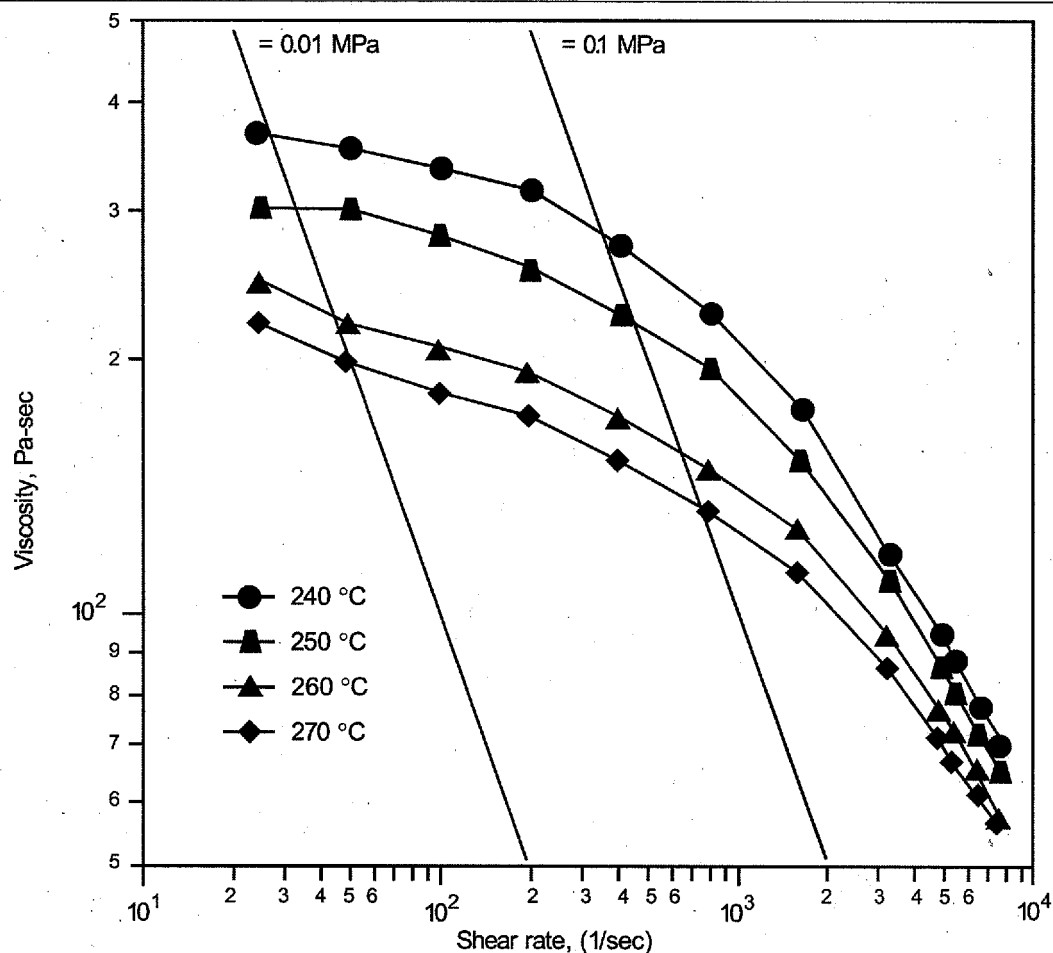


Figure 11/Melt Viscosity vs. Shear Rate.



Melt viscosity versus shear rate measured at four temperatures for a more recently made PTT polymer material, SH-2020 with about the same LVN, 0.92, as the previous material. These tests were done with the maximum assurance that all environmental conditions meet those required to eliminate water and excess oxygen as agents of degradation. Although the sample appears to be better behaved, there is still a marked departure from the parabolic curve shape with onset near to 260 °C and continuing at 270 °C.

Figure 12/PTT Extrusion.

Drying: No crystallization step
Close-loop hot-air dryer at 130 °C for 4 hours,
dew point preferably lower than -40 °C
Dry to <50 ppm
Desiccant: 13X with regular maintenance

Typical Melt Temperature: 250-265 °C

Figure 13/PTT Extrusion Conditions.

Extruder Temperatures (°C):

	Extruder					Adaptor			Die			Melt.Temp
	Zone 1	2	3	4	5	1	2	3	1	2	3	
PTT	230	250	260	265	260	250	250	250	245	245	245	260

Chill Roll Temperature: 10 – 15 °C Preferred

Stretching Temperatures:

MDO: 30 – 50 °C TDO: 45 – 60 °C

Stretching Ratio:

MDO: 3.0 – 3.5 TDO: 3.0 – 4.5

Figure 14/Physical and Electrical Properties on Un-filled ETP Polymers.

Physical Property	PET	PTT	PBT	Nyl 6,6 Dry	PC	Nyl 6 Dry
Tensile Strength, MPa	61.7	59.3	56.5	82.8	65.0	65
Flexural Modulus, GPa	3.11	2.76	2.34	2.83	2.35	2.2-2.7
HDT @ 1.8 MPa, °C	65	59	54	90	129	60
Notched Izod Impact, J/m	37	48	53	53	640	58.6
Specific Gravity	1.40	1.35	1.34	1.14	1.20	1.14
Mold Shrinkage, m/m	0.030	0.020	0.020	0.015	0.007	0.015
Melt Temperature, °C	265	225	228	265		230
Glass Trans. Temp., T _g , °C	80	45-75	25	50-90	150	50
Dielectric Strength, V/mil	550	530	400	600	380	380
Dielectric Constant 1MHz	3.0	3.0	3.1	3.1	3.6	3.3
Dissipation Factor 1 MHz	0.02	0.015	0.02	0.02	0.01	0.024
Volume Resistivity Ohm-cm	1.00 E+15	1.00 E+16	1.00 E+16	1.00 E+15	8.2 E+16	1.00 E+15

PTT data are reported from measurements made in our laboratory.
All other data are taken from the available polymer literature.

Figure 15/Tensile Properties ASTM D-882.

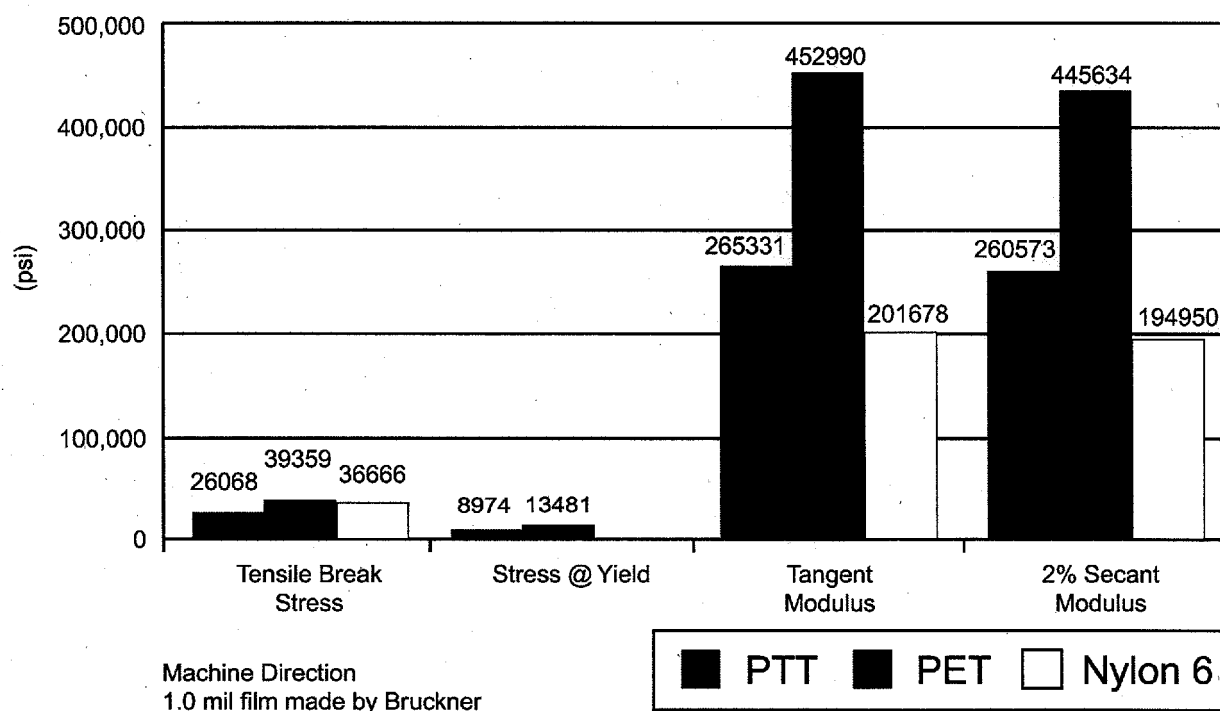


Figure 16/Shrinkage Force vs. Temperature ASTM D-2838.

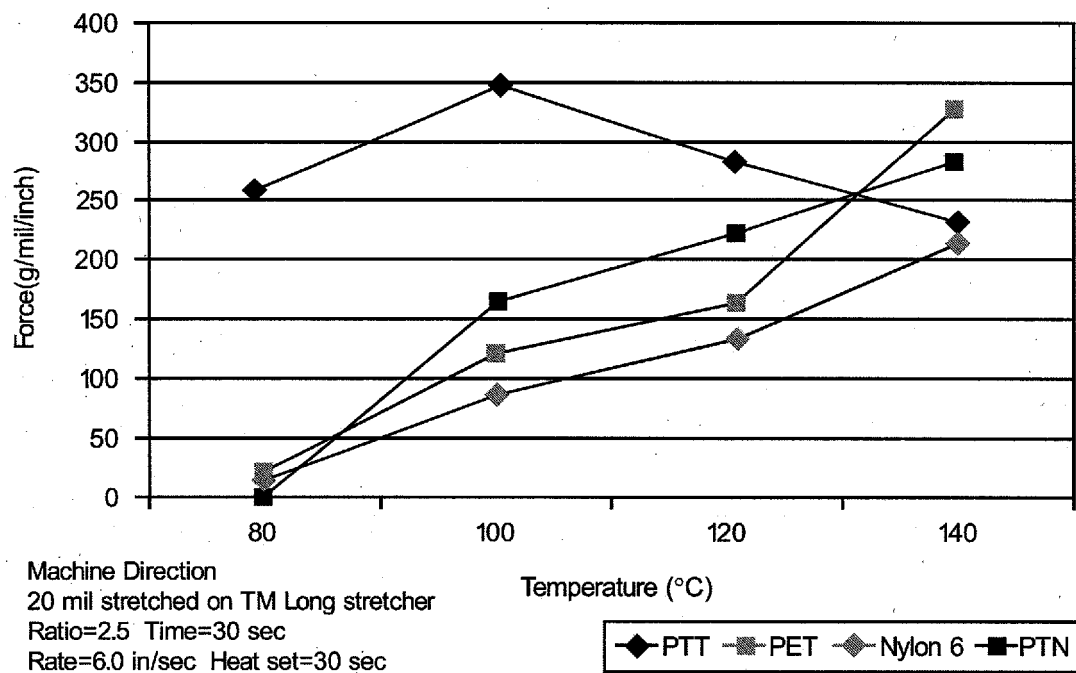


Figure 17/Percent Shrinkage vs. Temperature ASTM D-2732.

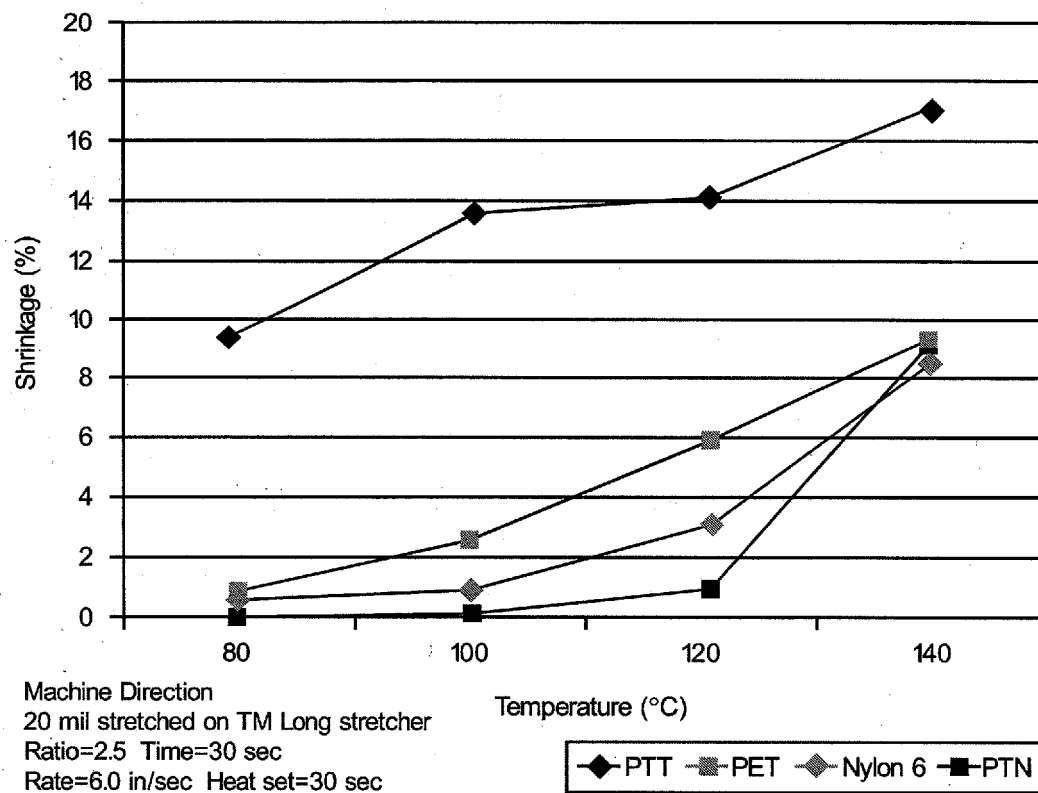
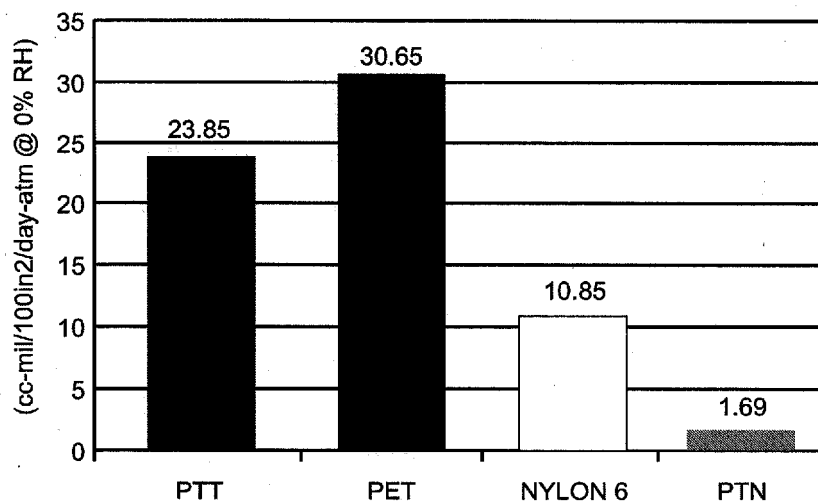
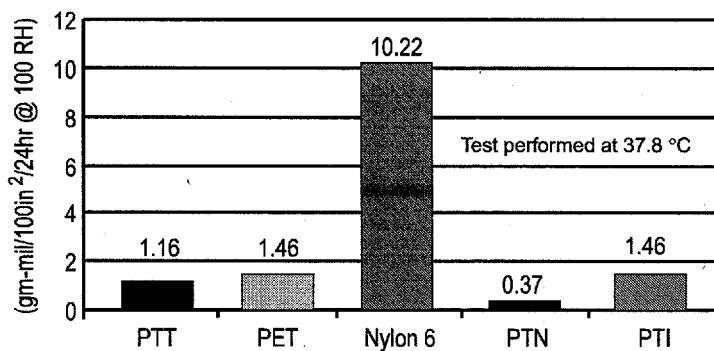


Figure 18/Carbon Dioxide Transmission Rate ASTM D-1434.



Test Performed by MOCON 1.0 mil film
PTT-PET-Nylon 6 made by Bruckner.
PTN made at WTC 20 mil stretched on TM Long stretcher

Figure 19/Water Vapor Transmission Rate ASTM F-1249.



20 mil stretched on TM Long stretcher Time=30 sec
Rate=6.0 in/sec Ratio=2.5 Heat Set=30 sec

Figure 20/Oxygen Transmission Rate ASTM D-3985.

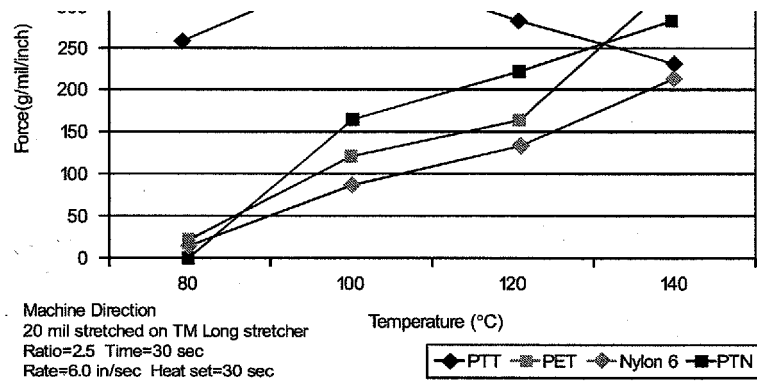
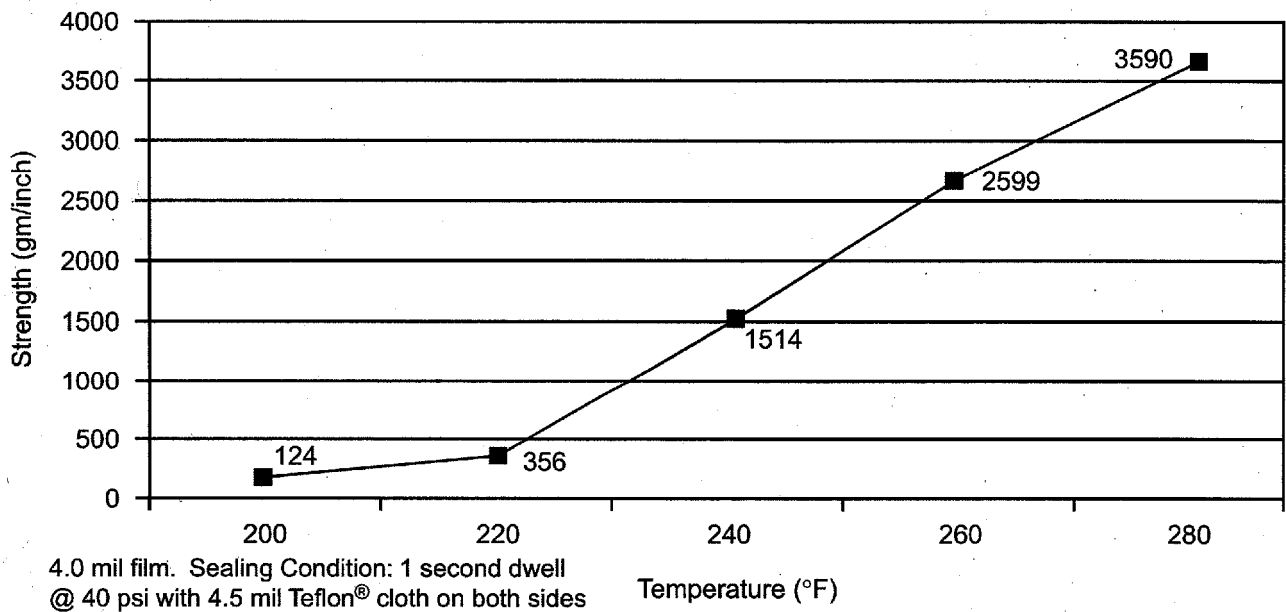


Figure 21/Seal Strength vs. Sealing Temperature of PTI Film.



For more information, please contact:

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